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Microstructure and piezoelectric properties of $Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}TiO_3-NaSbO_3$ ceramics

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ABSTRACT

Lead-free piezoelectric ceramics $(1-x)Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}TiO_3-xNaSbO_3$ have been prepared by a conventional ceramics technique, and their microstructure and electrical properties have been investigated. The addition of $NaSbO_3$ has no remarkable effect on the crystal structure within the studied doping content; however, an obvious change in microstructure took place. With increase in $NaSbO_3$ content, the temperature from a ferroelectric to antiferroelectric phase transition increases, and the temperature for a transition from antiferroelectric phases to paraelectric phases changes insignificantly. Simultaneously, the temperature range between the rhombohedral phase transition point and the Curie temperature point becomes smaller. The piezoelectric properties significantly increase with increase in $NaSbO_3$ content and the piezoelectric constant and electromechanical coupling factor attain maximum values of $d_{33}=160$ pC/N and $k_p=0.333$ at $x=0.01$. The results indicate that $(1-x)Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}TiO_3-xNaSbO_3$ ceramic is a promising lead-free piezoelectric candidate material.

1. Introduction

Lead-based piezoelectric ceramics, represented by $Pb(Zr,Ti)O_3$, $Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbTiO_3$, and $Pb(Zn_{1/3}Nb_{2/3})O_3$–$PbTiO_3$, have been widely applied in industry as sensor, actuator, and transducer materials due to their excellent electrical properties. However, the toxicity of lead oxide has been considered to be a serious threat to the environment and general public health. Therefore, there is a great need to develop lead-free ceramics with good piezoelectric properties for replacing the lead-based ceramics in various applications.

One of the most studied compounds of lead-free piezoelectric ceramics is ferroelectric bismuth sodium titanate $(Bi_{0.5}Na_{0.5})TiO_3$ (BNT), discovered by Smolenskii and Aganovskaya [1]. BNT has been considered a good candidate of lead-free piezoelectric ceramics because of its strong ferroelectricity at room temperature and high Curie temperature T_c of 320 °C [2–6]. However, pure BNT ceramics are difficult to pole because of their high coercive field ($E_c=7.3$ kV/mm). As compared to PZT ceramics, the pure BNT ceramics usually exhibit relatively weak piezoelectric properties ($d_{33}=83$ pC/N) [7]. In order to improve the poling process and enhance the piezoelectric properties of the BNT-based system, a number of BNT-based solid solutions, such as $(Bi_{0.5}Na_{0.5})_{(1-1.5x)}La_xTiO_3$[8], $Bi_{0.5}(Na_{1-x-y}K_xLi_y)_{0.5}TiO_3$ [9], $(Bi_{1/2}Na_{1/2})Ti_{1-x}(Zn_{1/3}Nb_{2/3})_xO_3$ [10], BNT–$Bi(Mg_{2/3}Nb_{1/3})O_3$ [11] and BNT–Ba(Ti, Zr)O_3 [12], have been developed and studied. Among the solid solutions that have been developed so far, $(1-x)Bi_{0.5}Na_{0.5}TiO_3-xBi_{0.5}K_{0.5}TiO_3$ (BNT–BKT) system has attracted considerable attention, because of the existence of rhombohedral–tetragonal morphotropic phase boundary (MPB) near $x=0.18$, where their electromechanical properties are sharply enhanced at this boundary [13]. However, the piezoelectric properties of BNT–BKT system are not good enough for most practical uses. On the other hand, the formation of a new solid has been shown to be an effective method to improve the piezoelectric properties of the BNT ceramics. Therefore, it is anticipated that by developing a multi-component system, the piezoelectric properties of the BNT ceramics can be further enhanced. Moreover, it was reported that introduction of $LiSbO_3$ or $AgSbO_3$ into the $Na_{0.5}K_{0.5}NbO_3$ solid solution and $NaNbO_3$ or $LiNbO_3$ into BNT-BKT ceramics could increase evidently the d_{33}-piezoelectric coefficient and electromechanical coupling [14–16]. Therefore, it is possible to improve the piezoelectric properties of $NaSbO_3$-added BNT-based ceramics. In the present work, a new ternary solid solution of $(1-x)Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}TiO_3-xNaSbO_3$ ($(1-x)$BNKT$-x$NB) was prepared by a conventional ceramic technique, and the effect of $NaSbO_3$ addition on the crystal structures and electromechanical properties was investigated.

2. Experimental procedure

A conventional mixed oxide route was utilized to prepare $(1-x)Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}TiO_3$ ceramics ($x=0$, 0.005, 0.01, 0.015, 0.020).

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Reagent grade oxide or carbonate powders of Bi_2O_3, TiO_2, Sb_2O_3, K_2CO_3, and Na_2CO_3 from Guangdong Xilong Chemical Co., Ltd., were used as starting materials. The powders were ball-milled for 12 h and calcined at 800–900 °C for 2 h. After calcination, the mixture was ball-milled for 24 h, dried, and granulated with PVA as a binder. The granulated powders were pressed into disks with diameter 18 mm and thickness 1.2 mm. The compacted disks were sintered at 1150 °C for 2 h in air. Silver paste was fired on both faces of the disks at 650 °C as electrodes. The specimens for measurement of piezoelectric properties were poled in DC-704 silicon oil from SCM Industrial Chemical Co., Ltd., at 40–50 °C under 3–4 kV/mm for 15 min.

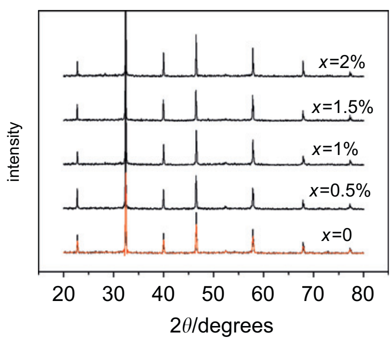

Fig. 1. XRD pattern of $(1-x)Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}TiO_3-xNaSbO_3$ ceramic.

The crystalline phase of sintered ceramics was identified by the X-ray diffractometer (Bruker D8-Advance) with Cu $K\alpha$ radiation ($\lambda = 1.5418$ Å) and graphite monochromater. The microstructure of sintered samples was observed by a scanning electron microscope (JSM–5610LV). Piezoelectric properties were measured using an impedance analyzer (Agilent 4294A) by a resonant and anti-resonant method. Piezoelectric constant d_{33} was measured by means of a quasi-static d_{33} meter (ZJ-3A, China) based on the Berlincourt method at 110 Hz. The dielectric properties were investigated using an impedance analyzer (Agilent 4294A).

3. Result and discussion

The XRD patterns of $(1-x)Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}TiO_3-xNaSbO_3$ ceramics are shown in Fig. 1. Similar to the BNT ceramics, all the $(1-x)Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}TiO_3$ $-xNaSbO_3$ ceramics possess a single-phase perovskite structure with rhombohedral symmetry and no secondary phase is observed because all reflections can be assigned to perovskite PSN (JCPDS 86-0225). The lattice parameters α and θ vary from 3.886 to 3.904 nm and from 89.56° to 89.64°. This suggests that $NaSbO_3$ has diffused into the BNT lattice to form a new solid solution, with Na^+ entering the $(Bi_{0.5}Na_{0.5})^{2+}$ sites and Sb^{5+} occupying the Ti^{4+} sites.

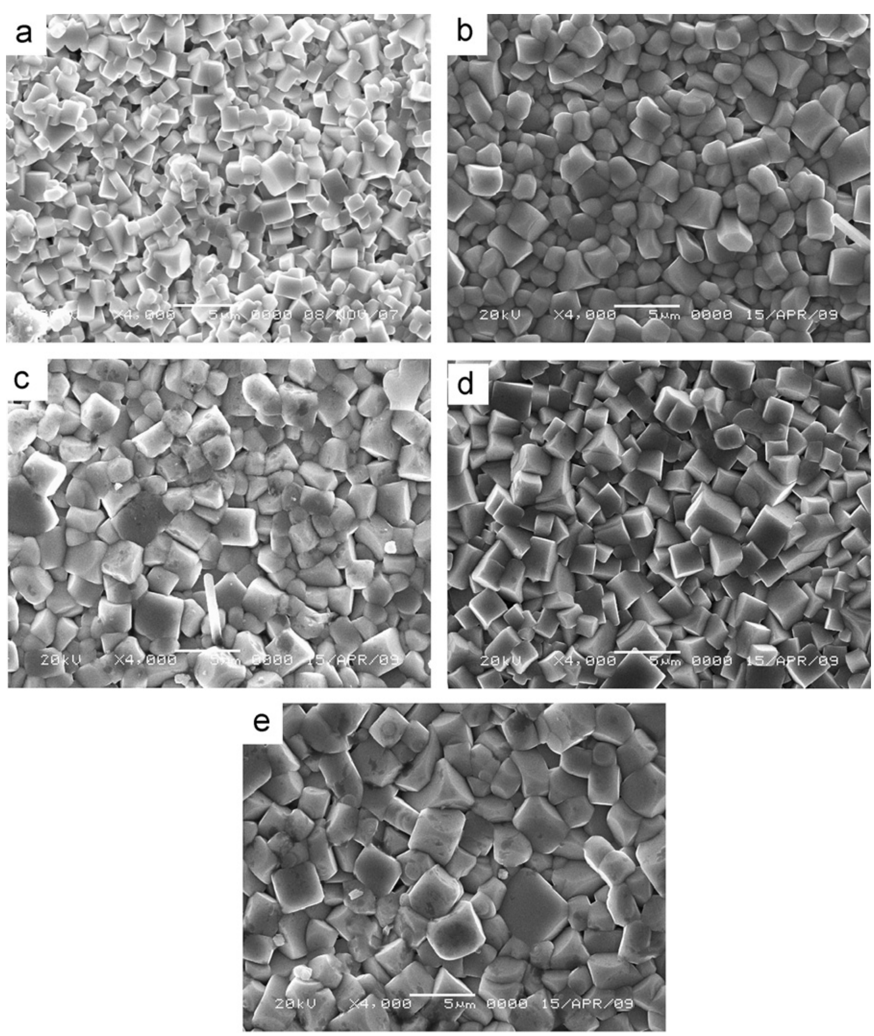

Fig. 2. SEM images of $(1-x)Bi_{0.5}(Na_{0.82}K_{0.18})_{0.5}TiO_3-xNaSbO_3$ ceramics: (a) $x=0$, (b) $x=0.005$, (c) $x=0.01$, (d) $x=0.0015$, and (e) $x=0.02$.